Prevention of Metallic Corrosion by Lauric Hydrazide and Its Salts Under Vapor Phase Conditions

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ABSTRACT: Six organic volatile corrosion inhibitors (VCI) were synthesized using lauric hydrazide with various acids such as cinnamic acid, succinic acid, nitrobenzoic acid, phthalic acid, and maleic acid and evaluated as corrosion inhibitors of mild steel, copper, brass, zinc, and aluminum by weight loss and potentiodynamic polarization methods. All the investigated VCI exhibited good inhibition efficiency for all the metals tested. The inhibition efficiency of all compounds increased as the inhibitor concentration increased. Lauric hydrazide cinnamate showed the best results among all compounds studied. In addition, all inhibitors showed anodic behavior. Adsorption studies demonstrated that all the investigated compounds followed Temkin's adsorption isotherm.

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Corrosion is defined as the destruction or deterioration of metals by chemical means in environments to which they are exposed (1). Corrosion causes enormous economic losses. In the United States, the economic losses have been estimated as high as \$270 billion per year. Corrosion of metal surfaces commonly occurs in the presence of oxygen and moisture and involves electrochemical reactions. Oxidation takes place at anodic sites and reduction occurs at cathodic sites. In an acidic medium, a hydrogen evolution reaction predominates, whereas in a neutral medium, reduction of oxygen takes place.

Corrosion inhibitors reduce or prevent these reactions. They are adsorbed onto the metal surface and act by forming a barrier to oxygen and moisture, by complexing with metal ions or by removing corrodants from the environment. Some of the inhibitors facilitate formation of a passivating film on the metal surface.

Volatile corrosion inhibitors (VCI) are used to protect metallic articles and equipment in an enclosed atmosphere (2). The choice of a chemical compound as a vapor phase VCI depends on its vapor pressure as well as its efficiency in preventing corrosion by forming a protective film. Numerous investigations of corrosion inhibition have utilized aliphatic amines, alicyclic amines, and their salts as VCI for various industrial metals and alloys (3–5). FA amines were more effective than

cyclic amines and aromatic amines (5). *m*-Dinitrobenzene with β-naphthol was examined as a VCI in a SO_2 and chloride atmosphere by Rajagopalan *et al.* (6). Subramanian *et al.* (7) studied the corrosion-inhibiting performance of cyclohexylamine salts and dicyclohexylamine salts on copper, mild steel, and zinc in an SO_2 environment. Dicyclohexylamine exhibited 70.86, 85.15, and 91.81% inhibition efficiency (IE) in mild steel, copper and zinc, respectively. Subramanian *et al.* (8) recently studied the corrosion-inhibiting behavior of morpholine and its three salts, i.e., morpholine carbonate, borate, and phosphate salts. Of these, morpholine and its carbonate salt exhibited 90 and 85% IE, respectively, whereas the other salts showed less than 40% IE.

Continuing our recent work (9–10) on FA derivatives as corrosion inhibitors, we report here the inhibiting properties of six organic vapor VCI, namely, lauric hydrazide (LH), lauric hydrazide cinnamate (LHC), lauric hydrazide nitrobenzoate (LHNB), lauric hydrazide maleate (LHM), lauric hydrazide succinate (LHS), and lauric hydrazide phthalate (LHP) on mild steel, brass, copper, aluminum, and zinc.

MATERIALS AND METHODS

Weight loss measurements. Corrosion experiments were carried out using various ferrous and nonferrous metals, *viz.,* mild steel, brass, copper, aluminum, and zinc. Specimens of size $2.0 \times 2.0 \times 0.025$ cm were used for weight loss measurement studies. Weight loss experiments were carried out in the presence and absence of inhibitors at concentrations ranging from 250–1000 ppm, using tight-fitting rubber-corked jars containing 25 mL of water. Concentrations of the inhibitors were taken according to the volume of the jar. Metal specimens were suspended in these bottles with nylon tags and just below these specimens, weighed VCI samples were kept in a glass container, to avoid contact with the liquid kept inside the jar, at a temperature of $35 \pm 1°C$ for 30 d. Relative humidity was kept at 100% to allow continuous condensation of moisture on the metal specimen.

Synthesis of inhibitors. LH was synthesized according to the procedure reported in the literature (11), and the salts were prepared by dissolving equimolar amounts of LH and organic acids in ethanol. The reaction mixture was stirred for 1.0 h at 40°C, and the precipitated compounds were filtered and crystallized from ethanol. Melting points (in °C) of the salts were as follows: LH = 85, LHC = 120, LHNB = 130, LHP = 135,

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TABLE 1
Name, Molecular Structure, and Vapor Pressure of Volatile Corrosion Inhibitors (VCI)

Serial no.	Molecular structure	Name	Vapor pressure (Pa)
1	$CH_3 \cdot (CH_2)_{10}$–$\overset{\overset{\text{O}}{\|\|}}{C}$–$NHNH_2$	Lauric hydrazide (LH)	1.87×10^{-4}
2	$CH_3 \cdot (CH_2)_{10}$–$\overset{\overset{\text{O}}{\|\|}}{C}$–$NHNH_2 \cdot HOOC$–$CH=CH$—⬡	Lauric hydrazide cinnamate (LHC)	0.80×10^{-4}
3	$CH_3 \cdot (CH_2)_{10}$–$\overset{\overset{\text{O}}{\|\|}}{C}$–$NHNH_2 \cdot HOOC$—⬡$O_2N$	Lauric hydrazide nitrobenzoate (LHNB)	1.23×10^{-4}
4	$CH_3 \cdot (CH_2)_{10}$–$\overset{\overset{\text{O}}{\|\|}}{C}$–$NHNH_2 \cdot HOOC$—⬡$HOOC$	Lauric hydrazide phthalate (LHP)	1.04×10^{-4}
5	$CH_3 \cdot (CH_2)_{10}$–$\overset{\overset{\text{O}}{\|\|}}{C}$–$NHNH_2 \cdot HOOC \cdot CH=CHCOOH$	Lauric hydrazide maleate (LHM)	1.54×10^{-4}
6	$CH_3 \cdot (CH_2)_{10}$–$\overset{\overset{\text{O}}{\|\|}}{C}$–$NHNH_2 \cdot HOOC \cdot CH_2$–$CH_2 \cdot COOH$	Lauric hydrazide succinate (LHS)	1.18×10^{-4}

LHM = 115, and LHS = 140. Names and molecular structures of these compounds are presented in Table 1.

Potentiodynamic polarization studies. For potentiodynamic polarization studies, metal strips without inhibitor and those with a 500 ppm concentration of inhibitor were embedded in araldite (a fixing material, made up of epoxy resin; Aldrich Chemical Co., St. Louis, MO) with an exposed area of 1.0 cm². Potentiodynamic polarization studies were carried out at a constant temperature of 28 ± 2°C according to ASTM methods G 3-74 and G 5-87 (12) using a potentiostat/galvanostat (model 173; EG&G, Gaithersburg, MD), a universal programmer (model 175, EG&G), and an X-Y recorder (model RE 0089; EG&G). A platinum foil was used as the auxiliary electrode and a saturated calomel electrode served as reference. Analyzed reagent-grade sodium sulfate (Na_2SO_4) (Merck) and double-distilled water were used for preparing test solutions of 1 N Na_2SO_4 for all experiments, which were used as electrolyte solutions.

Eschke test. Corrosion tests also were carried out according to the procedure reported in the literature (13) using mild steel, brass, copper, aluminum, and zinc. Polished strips of size $5.0 \times 2.0 \times 0.25$ cm were wrapped in a single layer of inhibitor-impregnated kraft paper and suspended in a climatic cabinet maintained at 90% relative humidity. One gram per square foot of each inhibitor was used for the experiment. The temperature cycle was set at 40°C for 12 h and at room temperature for another 12 h for condensation of the moisture. The duration of the test was 20 d. A similar experiment was done with metals covered with untreated kraft paper (control).

Determination of vapor pressure of the inhibitors. The weighed compound was placed in a glass container with a hole of 1.0 mm in diameter. The glass container was then placed in an oven set at 35°C for 30 d. Loss in mass was mea-

sured, and the values were substituted into the formula given below (14):

$$p = \frac{W}{At} \times \left(\frac{2\pi RT}{M} \right)^{1/2} \qquad [1]$$

where p = vapor pressure of the inhibitor, A = area of the hole in m², t = time in seconds, W = mass loss in kilograms, T = temperature in Kelvin, M = molecular mass of the compound, and R = gas constant (8.314 J K⁻¹ mol⁻¹). Values of the vapor pressures obtained are given in Table 1.

RESULTS AND DISCUSSION

Weight loss measurements. The values of IE values and corrosion rates obtained by weight loss methods at different LH salt concentrations at 35 ± 1°C are summarized in Table 2. IE and surface coverage (θ) were calculated using the following equations:

$$\% \text{ IE} = \left[\left(W^0 - W \right) / W^0 \right] \times 100 \qquad [2]$$

$$\theta = \left[\left(W^0 - W \right) / W^0 \right] \qquad [3]$$

where W^0 and W are the weight loss in the absence and presence of inhibitors, respectively. Table 2 indicates the increase in IE with increasing inhibitor concentration. The IE was obtained at 1,000 ppm. All VCI showed good IE, which may be attributed to the formation of a physical barrier between the metal and corrosive environment by the interaction of metal and inhibitor molecules. However, the difference in their inhibiting action can be explained by their molecular structure.

The corrosion-inhibiting action of LH is attributed to the
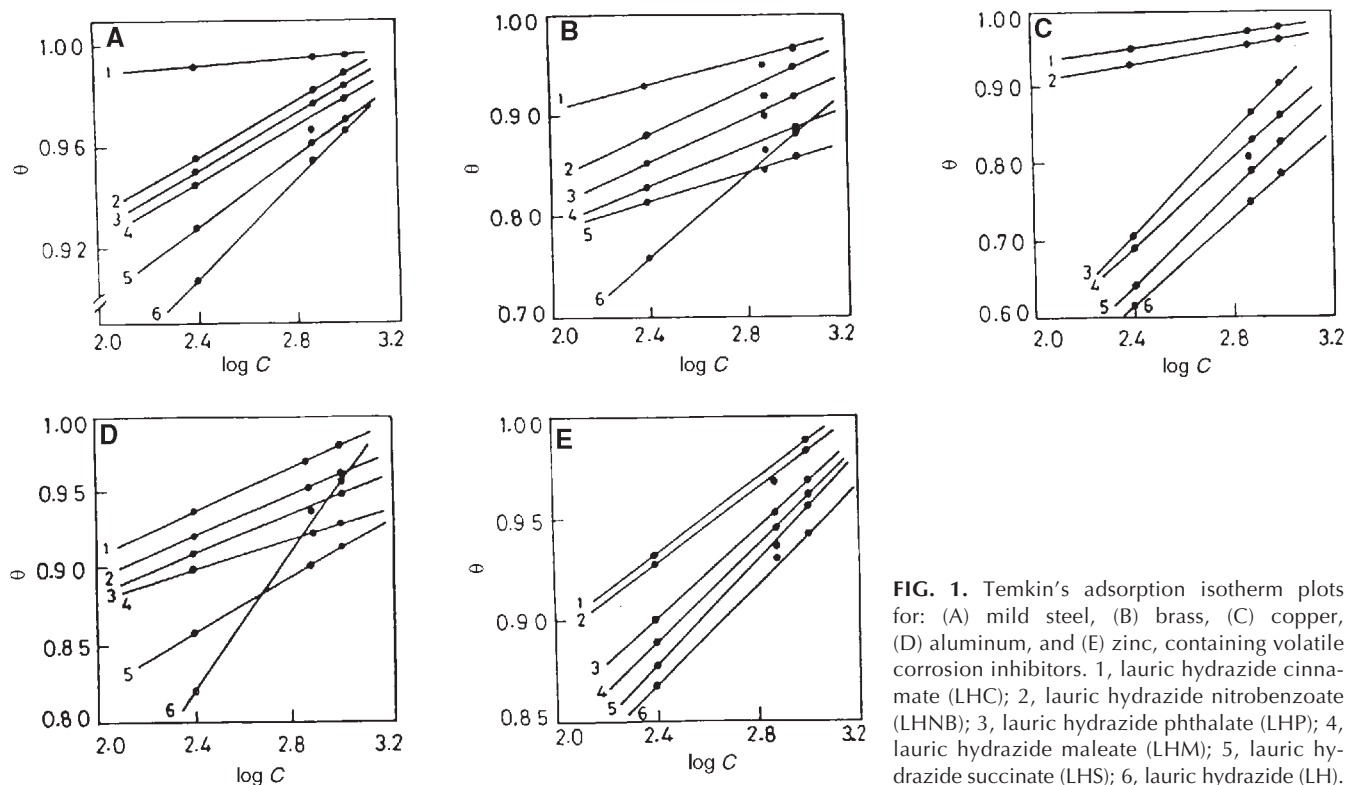
FIG. 1. Temkin's adsorption isotherm plots for: (A) mild steel, (B) brass, (C) copper, (D) aluminum, and (E) zinc, containing volatile corrosion inhibitors. 1, lauric hydrazide cinnamate (LHC); 2, lauric hydrazide nitrobenzoate (LHNB); 3, lauric hydrazide phthalate (LHP); 4, lauric hydrazide maleate (LHM); 5, lauric hydrazide succinate (LHS); 6, lauric hydrazide (LH).

presence of a lone pair of electrons on the N atom of the molecule that facilitates adsorption of inhibitor molecules on the metal surface. In the present investigation, the LHC salt exhibited good performance as a corrosion inhibitor owing to the presence of an additional π-bond between carbon atoms (–C=C–) which facilitates greater adsorption of cinnamate onto the metal surface, thereby giving high IE (16). The corrosion-inhibiting action of the LHNB is attributed to the presence of a nitro group. LHP showed better IE than the the LHM or LHS salts owing to the presence of an additional aromatic ring in the molecule. The inhibitive action of LHM is attributed to the presence of a double bond between the –C=C– atoms through which they adsorb strongly onto the surface of metals.

The degree of surface coverage (θ) for different inhibitor concentrations has been evaluated from weight loss values. The data were tested graphically by fitting to various isotherms. A straight line was obtained on plotting θ vs. log C (Figs. 1A–E), suggesting that the adsorption of LH salts onto the metal surface occurred according to Temkin's adsorption isotherm.

Eschke test. Visual observations of the metal specimens of this test are summarized in Table 3. The IE values and corrosion rates obtained in the presence and absence of various inhibitors are also summarized in Table 3.

Potentiodynamic polarization studies. Potentiodynamic polarization studies were carried out by immersing metal coupons and inhibited coupons in a 1 N Na_2SO_4 solution. The corrosion parameters, such as corrosion current density (I_{corr}), corrosion potential (E_{corr}), and percentage of IE (% IE) obtained from the potentiodynamic polarization curves are given in Table 2. The I_{corr} value was found to decrease significantly in the presence of the inhibitors, indicating that the compounds were effective corrosion inhibitors. All of these compounds shifted the E_{corr} in a more positive direction, suggesting that they were predominantly anodic inhibitors. Figures 2A–E show the Tafel plots obtained from different inhibited coupons in the presence of the 1 N Na_2SO_4 electrolyte solution for mild steel, brass, copper, aluminum, and zinc.

Mechanism of corrosion inhibition. Inhibition of metallic corrosion in the presence of LH salts involves vaporization of the inhibitor in a nondissociated molecular form, followed by hydrolysis of the salts into carboxylate anions (RCOO⁻) and organic cations (RCONHNH$_3^+$). Anions are adsorbed onto the anodic site of the metal and inhibit anodic reaction, whereas organic cations are adsorbed onto the cathodic site, thereby preventing cathodic reaction (17).

ACKNOWLEDGMENT

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TABLE 2
Weight Loss Parameters and Electrochemical Parameters Obtained from Different Vapor Phase VCI Concentrations at 40°C and 100% Humidity for 30 d for Mild Steel, Brass, Copper, Aluminum, and Zinc[a]

Inhibitor concentration (ppm)	Weight loss studies			Electrochemical studies		
	Weight loss (mg)	IE (%)	CR (mmpy $\times 10^{-2}$)	E_{corr} (mV)	I_{corr} (mA cm^2)	IE (%)
Mild steel						
Blank	38.5	—	5.95	−562	0.25	—
LH						
250	3.6	90.65	0.557	−448	0.042	83.20
750	1.8	95.32	0.279	−458	0.036	85.60
1,000	1.2	96.88	0.186	−430	0.032	87.20
LHC						
250	0.3	99.22	0.464	−426	0.014	94.40
750	0.2	99.48	0.310	−444	0.012	95.20
1,000	0.1	99.74	0.155	−416	0.008	96.80
LHNB						
250	1.7	95.58	2.63	−432	0.022	91.20
750	0.7	98.28	1.08	−428	0.018	92.80
1,000	0.4	98.96	0.62	−408	0.010	96.00
LHP						
250	1.9	95.06	2.94	−432	0.024	90.40
750	1.0	97.40	1.55	−446	0.022	91.20
1,000	0.6	98.44	1.29	−450	0.018	92.80
LHM						
250	2.1	94.54	3.25	−458	0.038	84.80
750	1.4	96.36	2.16	−464	0.030	88.00
1,000	0.8	97.14	1.24	−488	0.020	92.00
LHS						
250	2.8	92.73	4.33	−466	0.040	84.00
750	1.5	96.10	2.30	−446	0.034	86.40
1,000	1.1	97.14	1.70	−480	0.026	89.60
Brass						
Blank	12.0	—	1.72	−340	0.20	—
LH						
250	2.8	76.67	0.402	−290	0.050	75.00
750	1.6	86.33	0.230	−300	0.036	82.00
1,000	1.4	88.33	0.201	−270	0.026	87.00
LHC						
250	0.8	93.33	0.115	−260	0.020	90.00
750	0.6	95.00	0.086	−250	0.018	91.00
1,000	0.3	97.50	0.043	−222	0.010	95.00
LHNB						
250	1.4	88.33	0.201	−256	0.026	87.00
750	0.9	92.50	0.129	−254	0.024	88.64
1,000	0.5	95.83	0.718	−250	0.016	90.00
LHP						
250	2.0	83.33	0.287	−240	0.030	85.00
750	1.7	88.40	0.244	−252	0.022	89.00
1,000	0.7	89.20	0.101	−260	0.014	93.00
LHM						
250	1.8	85.00	0.259	−264	0.038	81.00
750	1.2	90.00	0.172	−270	0.028	86.00
1,000	0.9	92.50	0.129	−280	0.018	91.00
LHS						
250	2.1	82.53	0.302	−250	0.040	80.00
750	1.9	84.17	0.273	−240	0.032	84.00
1,000	1.2	86.74	0.172	−236	0.024	88.00
Copper						
Blank	9.7	—	1.317	−190	0.16	—
LH						
250	3.8	60.82	0.516	−160	0.048	70.00
750	2.7	72.16	0.367	−166	0.032	80.00
1,000	2.0	79.38	0.272	−150	0.024	85.00
LHC						
250	0.4	95.88	0.054	−140	0.028	82.50
750	0.3	96.90	0.041	−150	0.020	87.50
1,000	0.1	98.97	0.014	−112	0.012	92.50
LHNB						
250	0.6	93.81	0.081	−122	0.030	81.25
750	0.4	95.88	0.054	−138	0.024	85.00
1,000	0.2	97.94	0.027	−98	0.014	91.25

TABLE 2 (continued)

Inhibitor concentration (ppm)	Weight loss studies			Electrochemical studies		
	Weight loss (mg)	IE (%)	CR (mmpy $\times 10^{-2}$)	E_{corr} (mV)	I_{corr} (mA cm^2)	IE (%)
LHP						
250	2.8	71.13	0.338	−120	0.034	74.56
750	1.3	83.51	0.176	−122	0.032	80.00
1,000	0.8	91.75	0.109	−124	0.024	85.00
LHM						
250	3.0	69.07	0.407	−140	0.036	77.50
750	1.8	81.44	0.244	−136	0.024	85.00
1,000	1.2	87.63	0.163	−100	0.018	88.75
LHS						
250	3.4	64.95	0.462	−170	0.040	75.00
750	2.0	79.38	0.272	−160	0.032	80.00
1,000	1.6	83.51	0.217	−165	0.020	87.50
Aluminum						
Blank	60.6	—	27.30	−680	0.30	—
LH						
250	7.8	87.12	3.51	−640	0.050	83.33
750	4.1	93.23	1.85	−630	0.040	86.67
1,000	3.5	94.22	1.58	−654	0.038	87.33
LHC						
250	3.8	93.73	1.71	−610	0.028	90.67
750	1.4	97.69	0.631	−600	0.022	92.67
1,000	0.8	98.68	0.360	−595	0.018	94.00
LHNB						
250	4.2	93.07	1.89	−646	0.036	88.00
750	1.8	97.03	0.81	−638	0.028	90.67
1,000	1.2	98.02	0.541	−648	0.022	92.67
LHP						
250	5.8	90.43	2.61	−650	0.034	90.00
750	3.0	95.02	1.35	−646	0.030	92.68
1,000	1.6	97.36	0.721	−640	0.020	96.60
LHM						
250	6.4	89.44	2.88	−650	0.044	85.33
750	3.4	94.39	1.80	−640	0.038	87.33
1,000	2.1	96.53	0.946	−628	0.028	90.67
LHS						
250	7.0	88.45	3.15	−666	0.048	84.00
750	3.7	93.89	1.67	−642	0.040	86.67
1,000	2.2	96.37	0.991	−610	0.030	90.00
Zinc						
Blank	78.9	—	134.64	−708	0.38	—
LH						
250	14.0	82.26	2.39	−680	0.058	84.74
750	5.5	93.03	0.939	−676	0.048	87.37
1,000	3.2	95.94	0.546	−682	0.044	88.42
LHC						
250	4.5	94.30	0.768	−640	0.038	90.00
750	2.2	97.21	0.375	−632	0.026	93.16
1,000	1.5	98.10	0.256	−616	0.020	94.74
LHNB						
250	5.9	92.52	1.01	−658	0.042	88.95
750	3.8	95.10	0.648	−640	0.036	90.53
1,000	3.0	96.20	0.512	−638	0.024	93.68
LHP						
250	6.4	91.80	1.09	−666	0.048	87.37
750	5.6	93.36	0.956	−650	0.040	89.47
1,000	3.5	95.56	0.597	−640	0.030	92.68
LHM						
250	6.9	91.25	1.18	−688	0.048	87.37
750	5.8	92.65	0.990	−672	0.038	90.00
1,000	5.5	93.03	0.939	−670	0.028	92.63
LHS						
250	10.7	86.44	1.83	−680	0.050	86.84
750	7.2	90.87	1.23	−660	0.040	89.47
1,000	5.4	93.16	0.921	−652	0.032	91.58

[a]IE, inhibition efficiency; CR, corrosion rate; E_{corr}, corrosion potential; I_{corr}, corrosion current density. For other abbreviations see Table 1.
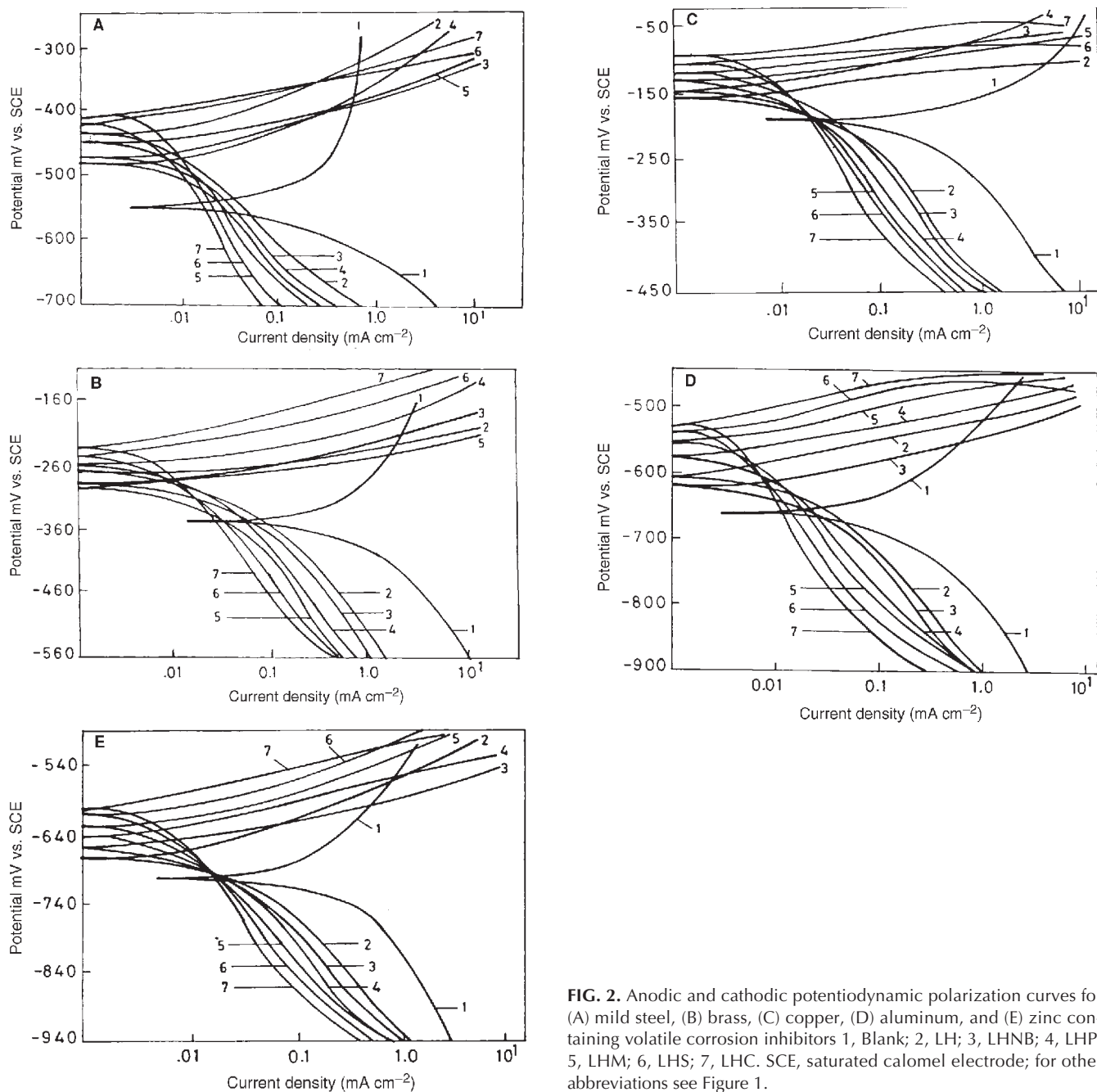
FIG. 2. Anodic and cathodic potentiodynamic polarization curves for (A) mild steel, (B) brass, (C) copper, (D) aluminum, and (E) zinc containing volatile corrosion inhibitors 1, Blank; 2, LH; 3, LHNB; 4, LHP; 5, LHM; 6, LHS; 7, LHC. SCE, saturated calomel electrode; for other abbreviations see Figure 1.

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TABLE 3
Results Obtained from the Eschke Test[a]

Metal	VCI	Extent of corrosion	Surface layer color	IE (%)	CR (mmpy $\times 10^{-3}$)
Mild steel	Control	High	Brownish-red	—	3.22
	LH	Moderate	Blackish-grey	90.38	0.31
	LHC	Very low	Dull white	96.15	0.12
Brass	Control	High	Yellowish-brown	—	1.35
	LH	Low	Dull yellow	82.98	0.23
	LHC	Very low	Yellow	96.43	0.13
Copper	Control	High	Reddish-brown	—	1.06
	LH	Moderate	Yellowish-brown	87.18	0.14
	LHC	Very low	Yellowish-brown	91.03	1.00
Bronze	Control	High	Deep brown	—	1.12
	LH	Low	Light brown	85.37	0.16
	LHC	Very low	Yellowish-brown	90.24	0.11
Aluminum	Control	High	Grayish-white	—	16.40
	LH	Moderate	Dull white	87.80	1.26
	LHC	Low	White	90.24	0.86

[a]For abbreviations see Tables 1 and 2.

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